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Title: Summary of DOE-EM Funded SRNL FY14 Activities Supporting the Joint EM-NE-SC International Study of Glass Behavior Over Geologic Time Scales

Introduction

The Savannah River National Laboratory (SRNL) participates in collaborative research activities with personnel from Pacific Northwest National Laboratory (PNNL), Argonne National Laboratory (ANL) and Oak Ridge National Laboratory (ORNL) involving the study of glass behavior over geologic time scales. The research areas span both modeling and experimental efforts and are funded by both DOE-EM and DOE-NE. The purpose of this memorandum is to summarize the FY14 DOE-EM funded activities performed by SRNL (HQTD1005, 2014).

Modeling Task

SRNL formulated a database comprised of numerous experimental long term glass dissolution studies in FY13 (Jantzen, 2013). This database entitled 'Accelerated Leach Testing of GLASS (ALTGLASS)' includes nine studies involving either both High Level Waste (HLW) glass surrogates or Low Activity Waste (LAW) surrogates totaling over 200 glasses. In FY14 several of the HLW data sets exhibiting either constant long-term rates or resumption of accelearted rates were examined in detail with respect to the leachate concentrations, the calculated gel concentrations and their relationship to the starting glass composition. A two-part manuscript has been submitted for review to the International Journal of Applied Glass Science (IJAGS) in September 2014. The first part paper entitled, 'Accelerated Leach Testingof GLASS (ALTGLASS): I. Waste Glass Hydrogel Compositions and Accelerated Dissolution'. The second paper

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is entitled '<u>Accelerated Leach Testing of GLASS</u>: II. Leachate-Hydrogel Interactions, Glass Structure, and Accelerated Dissolution'.

These manuscripts can be briefly summarized as follows. The durability of high level nuclear waste (HLW) glasses must be predicted on geological time scales. Predictive modeling approaches are currently based primarily on solution (leachate) concentrations. A compilation of long-term accelerated laboratory leach test data, known as ALTGLASS, was analytically interrogated by hierarchical modeling to determine how the {gel-leachate} interactions are controlled by the bulk glass composition and structure. Compositional differences were found between the hydrogels in glasses that resume accelerated dissolution (undesirable for long term performance) and those that do not resume accelerated dissolution (desirable for long term performance). Alkali (preferentially Li) was retained in the hydrogels of glasses that resumed accelerated dissolution and alkali was not retained to any great extent in the hydrogels of glasses that did not resume accelerated dissolution. Excess alkali in the glass appeared to cause the alkali retention in the hydrogel and caused excess strong base in the leachates to form as a function of time for the glasses that resumed accelerated dissolution. The excess strong base, [SB]_{ex}, in the leachate further reacts with the hydrogel to create the undesirable zeolite phases that are associated with glasses that resume accelerated dissolution. Glasses with no excess molar alkali did not resume accelerated leaching: the glass generated weak acids, [WA], in the leachate solution with time favoring gel aging into clays. Preliminary rate-determining leach layer forming exchange reactions were hypothesized.

Experimental Tasks

Product Consistency Test Interlaboratory Study

SRNL participated in an inter-laboatory powdered glass static leach testing study led by ANL aimed at generating data sets to access the variability involved in applying the ASTM Product Consistency Test to standard HLW surrogate glasses. Two different standard glasses, the Approved Reference Material (ARM-1) and a NIST Standard Reference Material (SRM-623) were used (Mellinger, 1983; Jantzen et al., 1995). Both glasses were prepared by ANL for the various laboratories by the protocol outlined in ASTM 1285C PCT-A which includes dry powder sieving followed by a final washing step with ultrapure ASTM Type I water and absolute ethanol to remove fines form the powders. Samples prepared in this way are designated 'd' for dry sieving. Data derived from this study will be compared to the acceptable range of PCT response using both unwashed and limited washed powders from early tests during the development of the PCT protocol in the 1987 to 1993 timeframe (Jantzen et al., 1995). SRNL also prepared both glasses using wet sieving methods using ultrapure ASTM Type I water and absolute ethanol for comparison to the dry sieving/washing steps. Samples prepared with wet sieving are designated 'w' for wet sieving. Another goal of this study was to compare the PCT-A response for these two glasses using smaller size fraction subsets of the typical (-)100/(+)200 mesh size. The smaller size fractions involved (-)100/(+)120, (-)120/(+)140 and (-)140/(+)200 sizes and were derived from resieving of a larger (-)100/(+)200 size fraction.

Leachate data from the SRNL testing is included in Appendix A. Table A-1 shows the as analyzed leachate concentrations. Filtered leachates were diluted using 6 mL of leachate to 4 mL of 0.4M ultrapure acid. The dilution corrected leachate concentrations are shown in Table A-2. Leachate concentrations were then normalized using the elemental mass fraction of each element for the SRM and the ARM glasses (See Jantzen et al., 1995) per Equation 1 below. These data are shown in Table A-3.

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NC_i = C_i(\text{sample}) / f_i Equation 1 where: NC_i = \text{normalized concentration, g waste form/L leachant,} C_i(\text{sample}) = \text{concentration of element "}i" in the solution, g_i/L, f_i = \text{mass fraction of element "}i" in the unleached waste form (unitless).
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The NC_i values were used to calculate the normalized elemental mass loss per Equation 2 below. These data are shown in Table A-4. Data pertaining to the mass of glass and leachant used and the calculated surface areas and surface area to volume ratios are shown in Table A-5 for the SRM glass and Table A-6 for the ARM glass.

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NL_i = NC_i / SA/V Equation 2 where: NL_i = \text{normalized elemental mass loss, g waste form/} m^2, \\ NC_i = \text{normalized concentration, g waste form/} L \text{ leachant, } g_i/L \\ SA/V = \text{surface area divided by the leachate volume, } m^2/L
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Equation 3 below was used to calculate the SA/V term for these PCT-A tests that used 1 g of glass powder to 10 mL of leachant. Densities for the SRM glass (2.24 g/cc) and the ARM glass (2.75 g/cc) were experimentally determined by ANL personnel for this study. Mean geometric diameters for the various sieve fractions used are 112 μ for (-)100/(+)200, 137 μ for (-)100/(+)120, 115 μ for (-)120/(+)140 and 89.5 μ for (-)140/(+)200.

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SA/V = (6 \text{ x mass}) / (\rho \text{ x d}) Equation 3 where: mass = mass \text{ of glass powder, g} \rho = glass \text{ density} d = mean geometric diameter for each glass powder fraction}
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Both the (-)100/(+)200 mesh size fractions of the dry sieved ARM and SRM glasses from ANL were analyzed for particle size distribution at SRNL. SRNL also analyzed the wet seived size fractions (all four distributions) for both the ARM and SRM glasses. The PSD measurements were performed on the glass powders using a Microtrac S3500 diffraction analysis instrument. This instrument quantifies particle diameters based on the portion of light scattered when a laser beam is projected through a fluid containing suspended solid phase particles. The amount and direction of light scattered by the particles is measured by an optical detector array and then analyzed to determine the PSD. The Microtrac S3500 instrument is

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capable of measuring particles with diameters between 0.24 to 1408 µm. PSD results were reported on a volume, number and area distribution basis. Volume distributions provide a means for assessing large particles, since large particles contribute most to the volume. In contrast, number distributions provide a means for evaluating small particles which can be abundant but contribute little to the volume. Mean particle diameters associated with the volume distributions and number distributions were also calculated and reported. The mean particle diameter associated with the volume distribution is denoted as 'mv' and represents the "center of gravity" of the distribution. This quantity is considered the average particle size contributing to the volume. The mean particle diameter associated with the number distribution is denoted as 'mn' and represents the average particle size contributing to the population. Details of relating PSD results measured on acircular particles relative to the mean distributions expected from sieve openings (geometric diameters) are discussed in a Microtrac reference document (Plantz, 2007).

Appendix B gives the summarized PSD results and some representative PSD distributions measured for the (-)100/(+)200 mesh size fractions of the dry sieved and wet sieved ARM glass.

The PCT data shown in Appendix A and all PSD data will be assessed by ANL personnel, along with data from other participating labs and presented at the upcoming 2014 Material Research Society meeting (Ebert, 2014).

Measurement of Viscosity for Internaional Standard Glass

SRNL led a program in 2012 to fabricate a six-component International Simple Glass (ISG) that could be used by United States and international researchers as a common glass for various studies related to the long term behavior over geologic time scales (Marra et al., 2012). Numerous 500-g blocks of this glass have been distributed world-wide for this purpose including past international collaborative workshops held at American Ceramic Society meetings of the Glass and Optical Materials Division (GOMD '12 (St. Louis), GOMD '13 (San Diego) and GOMD'14 in Aachen, Germany. SRNL received a request from PNNL personnel in 2014 for viscosity measurements on the ISG glass.

Viscosity measurements were performed on the ISG in FY2014 at SRNL using a Harrop high temperature viscometer as described in Schumacher et al., 2000. Initial measurements involved the sequence using a histeresis loop that started at the known processing melt temperature of the ISG glass at 1300 °C. The viscosity was measured in 25 °C increments up to 1400 °C, then measurements were made at 1300 °C in decreasing 25 °C increments through 1200 °C, followed by final measurements at 1156 °C and 1300 °C. Data for these measurements is shown in Table 1 and the measured viscosity vs. temperature is shown in Figure 1. A different method was also applied with a separate sample of the ISG glass that started viscosity measurements above the melt temperature at 1325 °C, with successive decreasing increments of 25 °C, down to a final temperature of 1125 °C. Data from the latter viscosity decreasing temperature method are shown in Table 2 and the viscosity vs. temperature plot is shown in Figure 2.

Table 1. Viscosity Data for Historesis Measurement Method

Sample	Rotation.	Brook.	Spindle	Meas.
Temp.	Speed	Torque	Const.	Viscosity
(C)	(RPM)	(%)	(K)	(Poise)
1302	60	28.9	96.02	46.25
1324	60	24.9	95.59	39.67
1348	60	21.2	95.12	33.61
1373	60	18.7	94.62	29.54
1400	60	16.6	94.09	26.03
1297	60	35.3	96.12	56.61
1277	60	41.9	96.52	67.40
1251	30	26.5	97.04	85.72
1227	30	33.1	97.51	107.70
1206	30	40.2	97.92	131.21
1156	12	27.2	98.92	223.93
1301	60	35.3	96.04	56.50

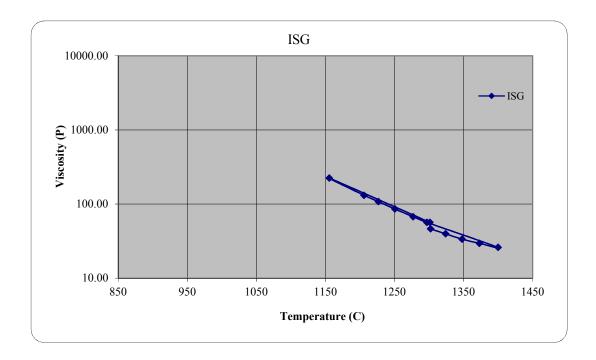


Figure 1. Plot of Viscosity vs. Temperature for the Historesis Method

Table 2. Viscosity Data for Decreasing Temperature Method

Sample	Rotation.	Brook.	Spindle	Meas.
Temp.	Speed	Torque	Const.	Viscosity
(C)	(RPM)	(%)	(K)	(Poise)
1324	30	12.8	95.59	40.89
1274	30	18.6	96.58	59.82
1225	30	28.1	97.55	91.42
1176	30	44.2	98.51	145.09
1125	12	30.9	99.52	255.99
1075	12	56.1	100.50	469.99
1024	6	59.1	101.52	999.39

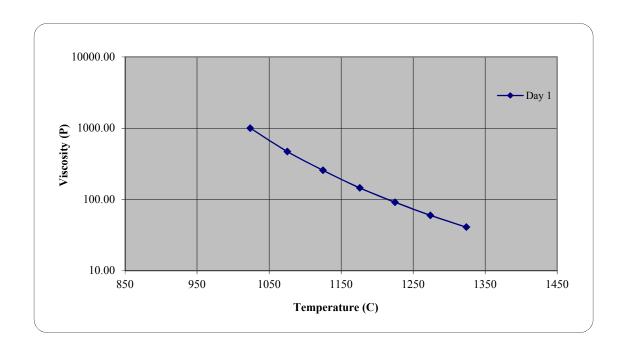


Figure 2. Plot of Viscosity vs. Temperature for the Decreasing Temperature Method

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SRNL-L3100-2014-00226, Rev. 0 Page 8 of 30 Appendix A **PCT Data**

Table A-1. As Analyzed PCT Leachate Concentrations, mg/L

	<u>Al</u>	<u>B</u>	<u>Ca</u>	<u>Li</u>	<u>Na</u>	<u>Si</u>
Blk-1	< 0.100	< 0.100	0.092	<1.00	<1.00	< 0.100
Blk-2	< 0.100	< 0.100	0.109	<1.00	<1.00	< 0.100
SRM-d-100/200-1	0.918	1.68	0.258	NA	3.94	17.9
SRM-d-100/200-2	1.09	1.74	0.211	NA	3.96	18.9
SRM-d-100/200-3	1.02	1.70	0.136	NA	3.92	18.9
SRM-w-100/200-1	0.967	1.83	0.182	NA	4.14	19.0
SRM-w-100/200-2	1.08	2.03	0.230	NA	4.39	20.5
SRM-w-100/200-3	1.11	2.00	0.177	NA	4.34	20.3
SRM-w-100/120-1	0.821	1.42	0.301	NA	3.42	16.4
SRM-w-100/120-2	0.866	1.43	0.239	NA	3.44	16.6
SRM-w-100/120-3	0.817	1.39	0.282	NA	3.45	16.4
SRM-w-120/140-1	0.893	1.59	0.147	NA	3.80	18.5
SRM-w-120/140-2	0.932	1.51	0.464	NA	3.77	17.4
SRM-w-120/140-3	0.920	1.58	0.202	NA	3.78	18.4
SRM-w-140/200-1	0.915	1.69	0.169	NA	4.04	19.1
SRM-w-140/200-2	0.955	1.67	0.197	NA	4.02	19.3
SRM-w-140/200-3	0.947	1.74	0.211	NA	4.01	19.3
ARM-d-100/200-1	2.46	8.15	0.160	6.80	18.5	34.5
ARM-d-100/200-2	2.49	8.53	0.127	6.96	19.0	35.2
ARM-d-100/200-3	2.48	8.74	0.188	7.08	19.3	35.5
ARM-w-100/200-1	2.83	8.96	0.436	7.65	19.9	35.1
ARM-w-100/200-2	2.69	9.39	0.192	7.81	20.4	36.0
ARM-w-100/200-3	2.73	9.58	0.253	8.01	21.0	37.2
ARM-w-100/120-1	2.72	8.13	0.154	6.59	17.7	32.7
ARM-w-100/120-2	2.46	8.06	0.161	6.60	17.8	32.6
ARM-w-100/120-3	3.80	7.84	0.151	6.48	17.4	32.1
ARM-w-120/140-1	2.63	8.91	0.295	7.35	19.5	35.2
ARM-w-120/140-2	2.54	8.63	0.141	7.21	19.1	34.4
ARM-w-120/140-3	2.43	8.60	0.107	7.08	18.5	33.4
ARM-w-140/200-1	3.58	9.76	0.188	8.36	21.5	36.8
ARM-w-140/200-2	2.88	10.3	0.129	8.53	22.2	38.1
ARM-w-140/200-3	2.94	10.6	0.271	8.81	23.0	39.7

Table A-2. Dilution Corrected PCT Leachate Concentrations, mg/L

	<u>Al</u>	<u>B</u>	<u>Ca</u>	<u>Li</u>	<u>Na</u>	<u>Si</u>
SRM-d-100/200-1	1.53	2.80	0.43	NA	6.57	29.87
SRM-d-100/200-2	1.81	2.91	0.35	NA	6.60	31.51
SRM-d-100/200-3	1.69	2.83	0.23	NA	6.54	31.46
SRM-w-100/200-1	1.61	3.05	0.30	NA	6.90	31.66
SRM-w-100/200-2	1.81	3.38	0.38	NA	7.31	34.23
SRM-w-100/200-3	1.85	3.33	0.29	NA	7.23	33.80
SRM-w-100/120-1	1.37	2.36	0.50	NA	5.69	27.25
SRM-w-100/120-2	1.44	2.39	0.40	NA	5.73	27.61
SRM-w-100/120-3	1.36	2.32	0.47	NA	5.74	27.31
SRM-w-120/140-1	1.49	2.65	0.24	NA	6.34	30.85
SRM-w-120/140-2	1.55	2.51	0.77	NA	6.28	29.00
SRM-w-120/140-3	1.53	2.63	0.34	NA	6.30	30.73
SRM-w-140/200-1	1.52	2.82	0.28	NA	6.73	31.89
SRM-w-140/200-2	1.59	2.79	0.33	NA	6.69	32.15
SRM-w-140/200-3	1.58	2.89	0.35	NA	6.68	32.23
ARM-d-100/200-1	4.09	13.58	0.27	11.34	30.83	57.53
ARM-d-100/200-2	4.15	14.22	0.21	11.59	31.71	58.66
ARM-d-100/200-3	4.14	14.57	0.31	11.80	32.22	59.13
ARM-w-100/200-1	4.71	14.93	0.73	12.75	33.23	58.42
ARM-w-100/200-2	4.48	15.65	0.32	13.02	34.01	59.93
ARM-w-100/200-3	4.55	15.97	0.42	13.35	35.06	61.93
ARM-w-100/120-1	4.53	13.55	0.26	10.98	29.50	54.51
ARM-w-100/120-2	4.10	13.43	0.27	10.99	29.73	54.37
ARM-w-100/120-3	6.34	13.07	0.25	10.80	28.98	53.51
ARM-w-120/140-1	4.39	14.85	0.49	12.25	32.44	58.70
ARM-w-120/140-2	4.23	14.39	0.24	12.01	31.86	57.38
ARM-w-120/140-3	4.05	14.34	0.18	11.79	30.83	55.60
ARM-w-140/200-1	5.97	16.26	0.31	13.93	35.82	61.35
ARM-w-140/200-2	4.80	17.10	0.22	14.22	36.95	63.50
ARM-w-140/200-3	4.91	17.69	0.45	14.69	38.35	66.11

Table A-3. Normalized Concentration Data, g/L

	Al	<u>B</u>	<u>Ca</u>	<u>Li</u>	<u>Na</u>	<u>Si</u>
SRM-d-100/200-1	4.59E-02	8.43E-02	8.61E-02	NA	1.38E-01	8.75E-02
SRM-d-100/200-2	5.43E-02	8.75E-02	7.03E-02	NA	1.39E-01	9.23E-02
SRM-d-100/200-3	5.07E-02	8.51E-02	4.54E-02	NA	1.38E-01	9.22E-02
SRM-w-100/200-1	4.83E-02	9.17E-02	6.07E-02	NA	1.45E-01	9.28E-02
SRM-w-100/200-2	5.42E-02	1.02E-01	7.65E-02	NA	1.54E-01	1.00E-01
SRM-w-100/200-3	5.56E-02	1.00E-01	5.88E-02	NA	1.52E-01	9.91E-02
SRM-w-100/120-1	4.10E-02	7.10E-02	1.00E-01	NA	1.20E-01	7.99E-02
SRM-w-100/120-2	4.33E-02	7.19E-02	7.96E-02	NA	1.21E-01	8.09E-02
SRM-w-100/120-3	4.09E-02	6.99E-02	9.39E-02	NA	1.21E-01	8.00E-02
SRM-w-120/140-1	4.46E-02	7.98E-02	4.89E-02	NA	1.33E-01	9.04E-02
SRM-w-120/140-2	4.66E-02	7.56E-02	1.54E-01	NA	1.32E-01	8.50E-02
SRM-w-120/140-3	4.60E-02	7.92E-02	6.73E-02	NA	1.33E-01	9.01E-02
SRM-w-140/200-1	4.57E-02	8.49E-02	5.63E-02	NA	1.42E-01	9.34E-02
SRM-w-140/200-2	4.77E-02	8.39E-02	6.58E-02	NA	1.41E-01	9.42E-02
SRM-w-140/200-3	4.73E-02	8.71E-02	7.03E-02	NA	1.41E-01	9.44E-02
fi SRM Glass	3.33	3.323	0.50	NA	4.75	34.12
ARM-d-100/200-1	1.38E-01	3.87E-01	1.67E-02	4.81E-01	4.30E-01	2.65E-01
ARM-d-100/200-2	1.40E-01	4.05E-01	1.33E-02	4.91E-01	4.42E-01	2.70E-01
ARM-d-100/200-3	1.40E-01	4.15E-01	1.97E-02	5.00E-01	4.49E-01	2.72E-01
ARM-w-100/200-1	1.59E-01	4.25E-01	4.56E-02	5.40E-01	4.63E-01	2.69E-01
ARM-w-100/200-2	1.51E-01	4.46E-01	2.01E-02	5.52E-01	4.74E-01	2.76E-01
ARM-w-100/200-3	1.54E-01	4.55E-01	2.64E-02	5.66E-01	4.89E-01	2.85E-01
ARM-w-100/120-1	1.53E-01	3.86E-01	1.61E-02	4.65E-01	4.11E-01	2.51E-01
ARM-w-100/120-2	1.38E-01	3.83E-01	1.68E-02	4.66E-01	4.14E-01	2.50E-01
ARM-w-100/120-3	2.14E-01	3.73E-01	1.58E-02	4.58E-01	4.04E-01	2.46E-01
ARM-w-120/140-1	1.48E-01	4.23E-01	3.08E-02	5.19E-01	4.52E-01	2.70E-01
ARM-w-120/140-2	1.43E-01	4.10E-01	1.48E-02	5.09E-01	4.44E-01	2.64E-01
ARM-w-120/140-3	1.37E-01	4.09E-01	1.12E-02	5.00E-01	4.30E-01	2.56E-01
ARM-w-140/200-1	2.02E-01	4.63E-01	1.97E-02	5.90E-01	4.99E-01	2.82E-01
ARM-w-140/200-2	1.62E-01	4.87E-01	1.35E-02	6.02E-01	5.15E-01	2.92E-01
ARM-w-140/200-3	1.66E-01	5.04E-01	2.84E-02	6.22E-01	5.35E-01	3.04E-01

Table A-4. Normalized Elemental Mass Loss Data, g/m²

	Al	<u>B</u>	<u>Ca</u>	<u>Li</u>	<u>Na</u>	<u>Si</u>
SRM-d-100/200-1	1.92E-02	3.52E-02	3.60E-02	NA	5.78E-02	3.66E-02
SRM-d-100/200-2	2.27E-02	3.65E-02	2.94E-02	NA	5.81E-02	3.86E-02
SRM-d-100/200-3	2.12E-02	3.56E-02	1.90E-02	NA	5.76E-02	3.86E-02
SRM-w-100/200-1	2.02E-02	3.83E-02	2.54E-02	NA	6.08E-02	3.88E-02
SRM-w-100/200-2	2.26E-02	4.25E-02	3.20E-02	NA	6.43E-02	4.19E-02
SRM-w-100/200-3	2.32E-02	4.18E-02	2.46E-02	NA	6.36E-02	4.14E-02
SRM-w-100/120-1	2.10E-02	3.63E-02	5.12E-02	NA	6.13E-02	4.08E-02
SRM-w-100/120-2	2.21E-02	3.67E-02	4.06E-02	NA	6.16E-02	4.13E-02
SRM-w-100/120-3	2.09E-02	3.58E-02	4.80E-02	NA	6.19E-02	4.09E-02
SRM-w-120/140-1	1.92E-02	3.43E-02	2.10E-02	NA	5.73E-02	3.88E-02
SRM-w-120/140-2	2.00E-02	3.25E-02	6.64E-02	NA	5.68E-02	3.65E-02
SRM-w-120/140-3	1.97E-02	3.40E-02	2.89E-02	NA	5.70E-02	3.87E-02
SRM-w-140/200-1	1.53E-02	2.84E-02	1.88E-02	NA	4.74E-02	3.12E-02
SRM-w-140/200-2	1.59E-02	2.80E-02	2.19E-02	NA	4.71E-02	3.14E-02
SRM-w-140/200-3	1.58E-02	2.91E-02	2.35E-02	NA	4.70E-02	3.15E-02
ARM-d-100/200-1	7.11E-02	1.99E-01	8.58E-03	2.47E-01	2.21E-01	1.36E-01
ARM-d-100/200-2	7.19E-02	2.08E-01	6.83E-03	2.52E-01	2.27E-01	1.38E-01
ARM-d-100/200-3	7.17E-02	2.13E-01	1.01E-02	2.56E-01	2.30E-01	1.39E-01
ARM-w-100/200-1	8.16E-02	2.18E-01	2.34E-02	2.77E-01	2.38E-01	1.38E-01
ARM-w-100/200-2	7.78E-02	2.29E-01	1.03E-02	2.83E-01	2.43E-01	1.42E-01
ARM-w-100/200-3	7.90E-02	2.34E-01	1.36E-02	2.91E-01	2.51E-01	1.46E-01
ARM-w-100/120-1	9.61E-02	2.42E-01	1.01E-02	2.92E-01	2.58E-01	1.57E-01
ARM-w-100/120-2	8.70E-02	2.40E-01	1.06E-02	2.93E-01	2.60E-01	1.57E-01
ARM-w-100/120-3	1.35E-01	2.34E-01	9.92E-03	2.87E-01	2.54E-01	1.55E-01
ARM-w-120/140-1	7.82E-02	2.23E-01	1.62E-02	2.74E-01	2.38E-01	1.42E-01
ARM-w-120/140-2	7.55E-02	2.16E-01	7.79E-03	2.69E-01	2.34E-01	1.39E-01
ARM-w-120/140-3	7.22E-02	2.15E-01	5.90E-03	2.63E-01	2.27E-01	1.35E-01
ARM-w-140/200-1	8.27E-02	1.90E-01	8.06E-03	2.42E-01	2.05E-01	1.16E-01
ARM-w-140/200-2	6.66E-02	2.00E-01	5.54E-03	2.47E-01	2.11E-01	1.20E-01
ARM-w-140/200-3	6.80E-02	2.07E-01	1.16E-02	2.55E-01	2.19E-01	1.25E-01

Table A-5. SRM PCT Data and Leachate pH

Sample ID	SRM-d-100/200-	SRM-d-100/200- 2	SRM-d-100/200- 3	SRM-w100/200-	SRM-w-100/200- 2	SRM-w-100/200-			
Vessel ID	288	318	319	320	362	366			
рН	8.76	8.79	8.79	8.97	9.10	9.05			
Empty mass (g)	123.341	122.737	120.422	122.946	122.805	123.417			
Mass w/ glass (g)	124.342	123.738	121.422	123.946	123.806	124.418			
Mass w/ water + glass (g)	134.342	133.739	131.424	133.947	133.808	134.417			
Mass (g)	1.001	1.001	1.000	1.000	1.001	1.001			
Vol. (cm ³)	10.000	10.001	10.002	10.001	10.002	9.999			
Vol./mass (cm ³ /g)	9.990	9.991	10.002	10.001	9.992	9.989			
SA, cm ²	239.4	239.4	239.2	239.2	239.4	239.4			
SA/V, m ⁻¹	2394.0	2393.7	2391.1	2391.3	2393.5	2394.2			
Sample ID	SRM-w-100/120 -1	SRM-w- 100/120-2	SRM-w-100/120- 3	SRM-w120/140- 1	SRM-w-120/140- 2	SRM-w-120/140- 3	SRM-w-140/200- 1	SRM-w-140/200- 2	SRM-w-140/200- 3
Vessel ID	368	370	B235	P93	P110	P112	P113	P114	P115
pН	8.65	8.82	8.68	8.59	8.63	8.71	8.81	8.76	8.81
Empty mass (g)	121.745	122.615	120.960	121.659	121.062	120.940	120.626	121.061	121.326
Mass w/ glass (g)	122.746	123.617	121.960	122.659	122.061	121.940	121.626	122.062	122.327
Mass w/ water + glass (g)	132.746	133.617	131.962	132.660	132.064	131.941	131.628	132.060	132.327
Mass (g)	1.001	1.002	1.000	1.000	0.999	1.000	1.000	1.001	1.001
Vol. (cm ³)	10.000	10.000	10.002	10.001	10.003	10.001	10.002	9.998	10.000
Vol./mass (cm ³ /g)	9.990	9.980	10.002	10.001	10.013	10.001	10.002	9.988	9.990
SA, cm ²	195.7	195.9	195.5	232.9	232.7	232.9	299.3	299.6	299.6
SA/V, m ⁻¹	1957.1	1959.1	1954.8	2329.0	2326.2	2329.0	2992.2	2996.4	2995.8

Table A-6. ARM PCT Data and Leachate pH

Sample ID	ARM-d-100/200-	ARM-d-100/200- 2	ARM-d-100/200- 3	ARM-w-100/200-	ARM-w-100/200- 2	ARM-w-100/200-			
Vessel ID	P124	P133	P144	P146	P151	P152			
рН	10.33	10.32	10.26	10.28	10.30	10.31			
Empty mass (g)	121.002	121.430	121.192	121.295	121.478	119.850			
Mass w/ glass (g)	122.001	122.431	122.193	122.296	122.478	120.850			
Mass w/ water + glass (g)	132.003	132.433	132.192	132.295	132.480	130.852			
Mass (g)	0.999	1.001	1.001	1.001	1.000	1.000			
Vol. (cm ³)	10.002	10.002	9.999	9.999	10.002	10.002			
Vol./mass (cm ³ /g)	10.012	9.992	9.989	9.989	10.002	10.002			
SA, cm ²	194.6	195.0	195.0	195.0	194.8	194.8			
SA/V, m ⁻¹	1945.7	1949.6	1950.2	1950.2	1947.7	1947.7			
	100/100	100/100	100/100	100/110	100/110	100/140	77.6	77.6	73.6
Sample ID	ARM-w-100/120- 1	ARM-w-100/120- 2	ARM-w-100/120- 3	ARM-w-120/140- 1	ARM-w-120/140- 2	ARM-w-120/140- 3	RM-w-140/200- 1	RM-w-140/200- 2	RM-w-140/200- 3
Vessel ID	P159	P183	P185	P186	P187	P188	P189	P190	P191
pН	10.24	10.31	10.29	10.37	10.33	10.30	10.33	10.37	10.37
Empty mass (g)	120.746	121.207	120.008	123.061	121.322	122.478	120.193	121.174	120.869
Mass w/ glass (g)	121.746	122.207	121.008	124.061	122.321	123.478	121.194	122.174	121.870
Mass w/ water + glass (g)	131.747	132.208	131.010	134.061	132.323	133.477	131.192	132.175	131.870
Mass (g)	1.000	1.000	1.000	1.000	0.999	1.000	1.001	1.000	1.001
Vol. (cm ³)	10.001	10.001	10.002	10.000	10.002	9.999	9.998	10.001	10.000
Vol./mass (cm ³ /g)	10.001	10.001	10.002	10.000	10.012	9.999	9.988	10.001	9.990
SA, cm ²	159.3	159.3	159.3	189.7	189.5	189.7	244.0	243.8	244.0
SA/V, m ⁻¹	1592.4	1592.4	1592.2	1897.2	1895.0	1897.4	2440.7	2437.5	2440.2

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Appendix B

Particle Size Distribution Data

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Tables B-1 and B-2 show the expected mean particle size for each sieve range along with the measured mean particle sizes from the various PSD analyses for the ARM and SRM glass powders, respectively. Each size fraction was analyzed using the volume distribution in both absorbance and transparent mode, followed by the number distribution and the area distribution. Very little difference was noted in the PSD data between the absorbance mode and transparent mode for any of the three distributions (Volume, Number or Area). The range of measured mean particle sizes for each measurement are compared to the geometric mean. These data show that both the mean particle sizes for both the volume and area distributions are higher than the geometric mean, whereas the number distribution mean particle sizes are in good agreement with the geometric mean. Example PSD data are shown for the dry sieved (-)100/(+)200 mesh size fractions ARM glass (six spectra/data sets summarized in the ANL ARM dry sieved row of Table B-1) as Figures B-1 thorugh B-6. Similar data for the wet sieved (-)100/(+)200 mesh size fractions ARM glass (six spectra/data sets summarized in the SRNL ARM wet sieved row of Table B-1) are shown in Figures B-7 through B-12. Plots of the '% Passing' vs. micron size for all of the volume and area distribution PSDs incidate a gaussian, single mode distribution for the dry sieved and washed, or the wet sieved preparations. These data indicate that either preparation method appears to adequately remove the fines generated in grinding of the glasses to produce powders for the static leach tests. Plots of the number distributions show a larger percentage passing at the lower particle sizes, i.e., slightly nongaussian, and an abrupt cut-off at the lower size tail of the distribution (Figures B-3, B-4 and Figures B-9 and B-10).

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Table B-1. PSD Data Summarized for ARM Glass

ANL - ARM		Volume	(Abs.&Trans.)	Number	(Abs.&Trans.)	Area	(Abs.&Trans.)
(dry sieve, wash)		50 Percentile Vol	Ratio	50 Percentile Num	Ratio	50 Percentile Area	Ratio
Mean (μ)	Mesh	Measured	Meas./Calc.	Measured	Meas./Calc.	Measured	Meas./Calc.
112	100/200	157 - 158	1.41	113-119	1.04	144-145	1.30
137	100/120	173 - 174	1.27	144-145	1.05	163-164	1.19
115	120/140	148	1.29	119-121	1.04	137-138	1.20
89.5	140/200	119-120	1.34	91-93	1.03	109-110	1.22
SRNL - ARM		Volume	(Abs.&Trans.)	Number	(Abs.&Trans.)	Area	(Abs.&Trans.)
(wet sieve, no wash)		50 Percentile Vol	Ratio	50 Percentile Num	Ratio	50 Percentile Area	Ratio
Mean (μ)		Measured	Meas./Calc.	Measured	Meas./Calc.	Measured	Meas./Calc.
112	100/200	151-152	1.36	107-113	0.99	138-140	1.25
137	100/120	174 - 175	1.27	144-145	1.05	163-164	1.19
115	120/140	150-151	1.31	121-122	1.06	140	1.22
89.5	140/200	119	1.33	91-93	1.03	109-110	1.22

Table B-2. PSD Data Summarized for SRM Glass

ANL - SRM		Volume	(Abs.&Trans.)	Number	(Abs.&Trans.)	Area	(Abs.&Trans.)
(dry sieve, wash)		50 Percentile Vol	Ratio	50 Percentile Num	Ratio	50 Percentile Area	Ratio
Mean (μ)		Measured	Meas./Calc.	Measured	Meas./Calc.	Measured	Meas./Calc.
112	100/200	150-152	1.35	111-116	1.02	138-140	1.25
NA	NA	NA	NA	NA	NA	NA	NA
NA	NA	NA	NA	NA	NA	NA	NA
NA	NA	NA	NA	NA	NA	NA	NA
SRNL - SRM		Volume	(Abs.&Trans.)	Number	(Abs.&Trans.)	Area	(Abs.&Trans.)
(wet sieve, no wash)		50 Percentile Vol	Ratio	50 Percentile Num	Ratio	50 Percentile Area	Ratio
Mean (μ)		Measured	Meas./Calc.	Measured	Meas./Calc.	Measured	Meas./Calc.
112	100/200	157	1.41	113-118	1.04	143-145	1.29
137	100/120	173-174	1.27	141-143	1.04	162-163	1.19
115	120/140	144-145	1.26	116-117	1.01	135-136	1.18
89.5	140/200	116-117	1.30	90-92	1.02	107-108	1.20

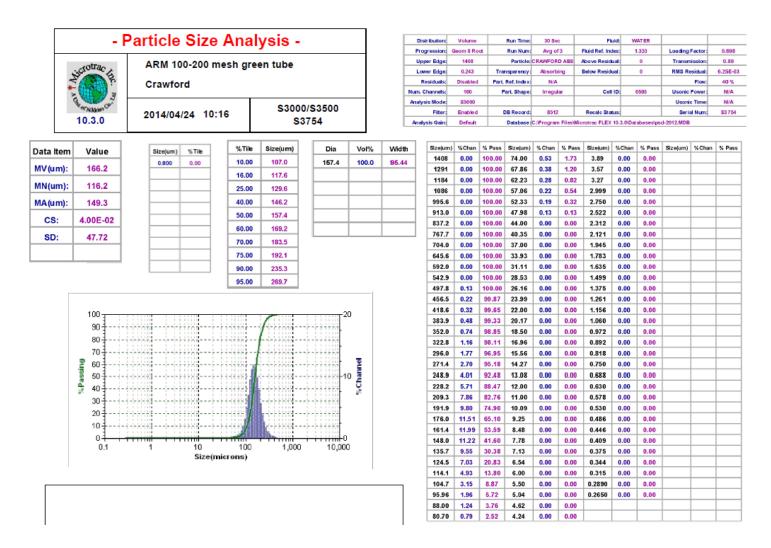


Figure B-1. ARM (-)100/(+)200 mesh, dry sieved, volume distribution, absorbance mode

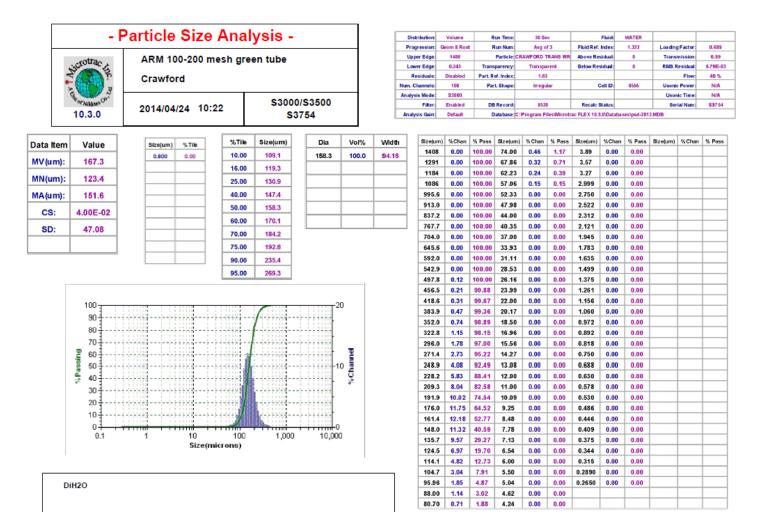


Figure B-2. ARM (-)100/(+)200 mesh, dry sieved, volume distribution, transparent mode

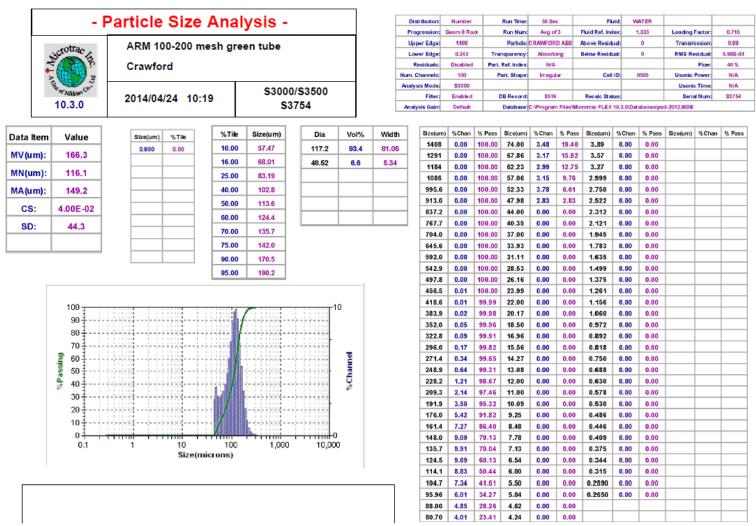


Figure B-3. ARM (-)100/(+)200 mesh, dry sieved, number distribution, absorbance mode

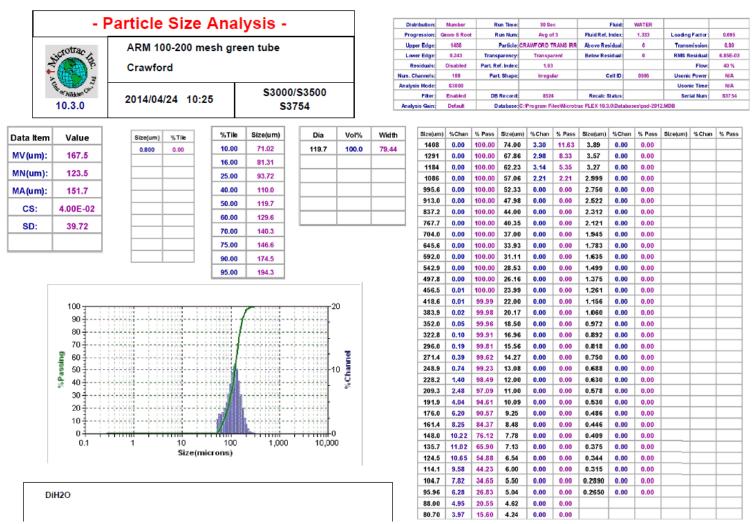


Figure B-4. ARM (-)100/(+)200 mesh, dry sieved, number distribution, transparent mode

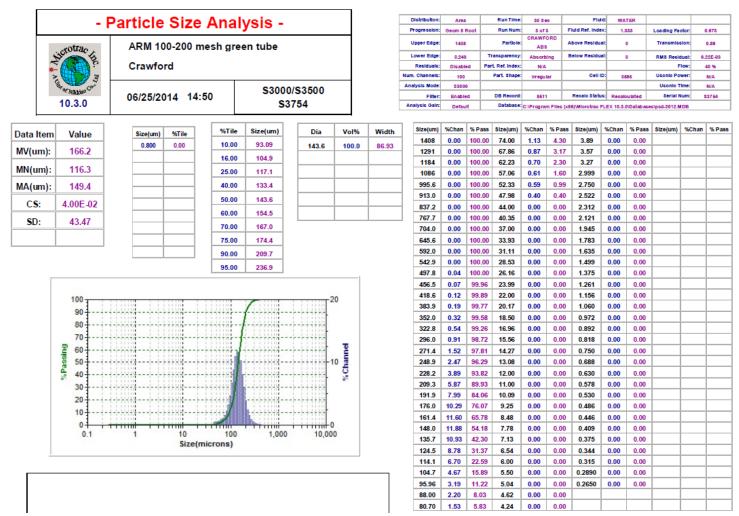


Figure B-5. ARM (-)100/(+)200 mesh, dry sieved, area distribution, absprbamce mode

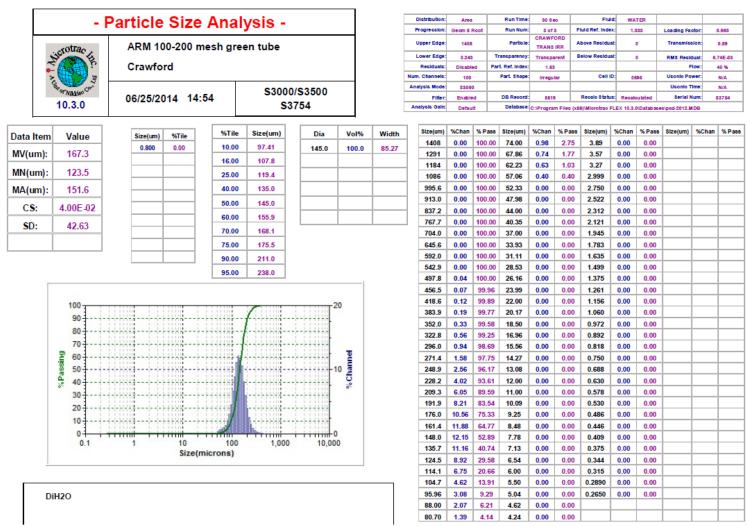


Figure B-6. ARM (-)100/(+)200 mesh, dry sieved, area distribution, transparent mode

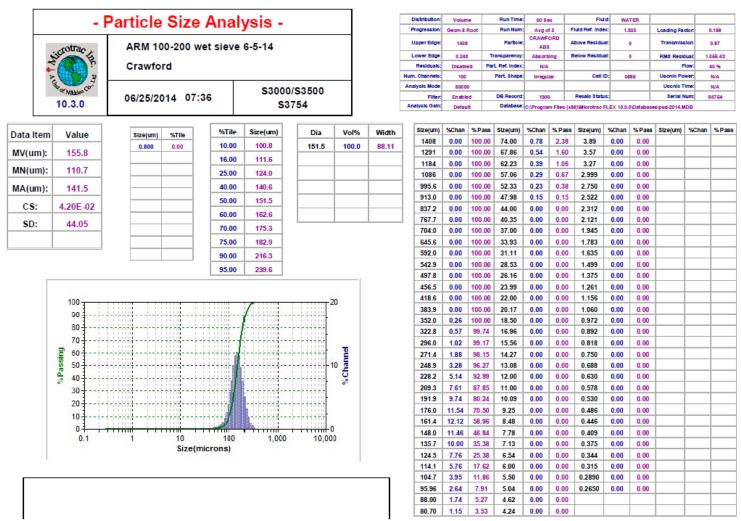


Figure B-7. ARM (-)100/(+)200 mesh, wet sieved, volume distribution, absorbance mode

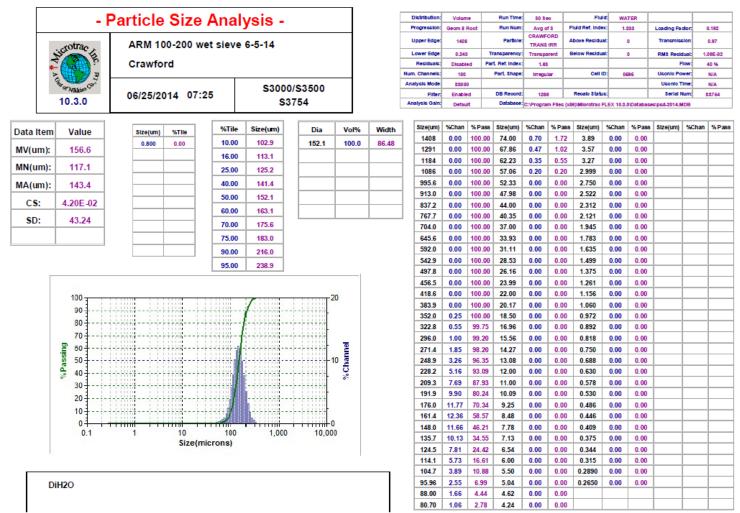


Figure B-8. ARM (-)100/(+)200 mesh, wet sieved, volume distribution, transparent mode

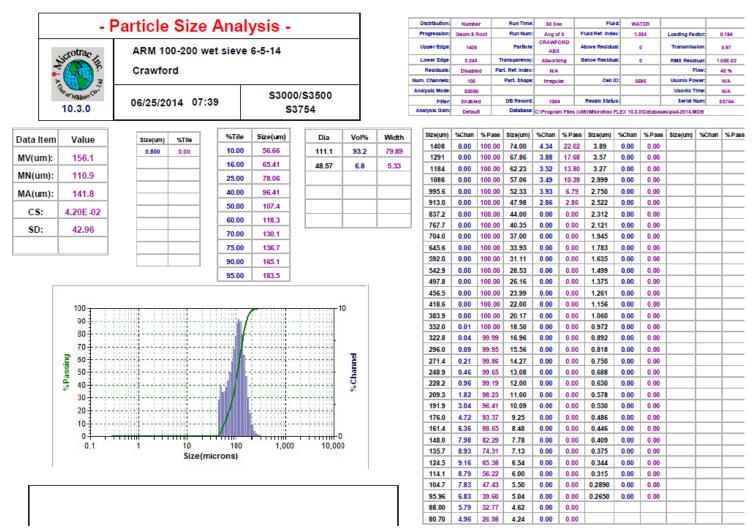


Figure B-9. ARM (-)100/(+)200 mesh, wet sieved, number distribution, absorbance mode

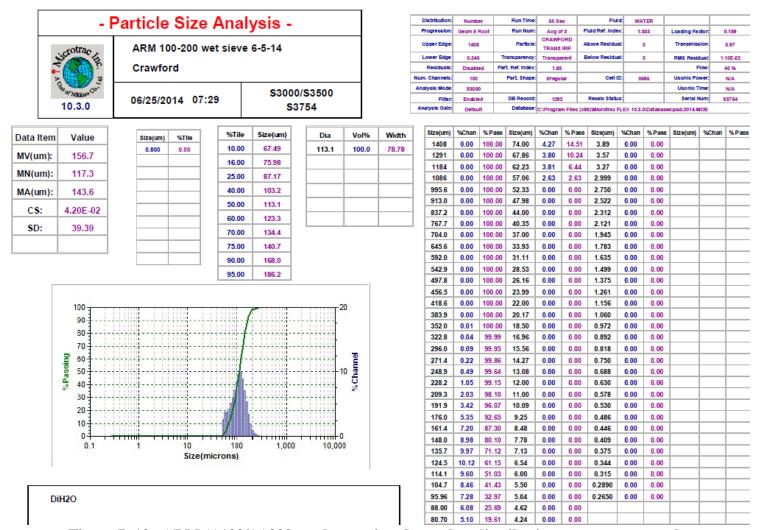


Figure B-10. ARM (-)100/(+)200 mesh, wet sieved, number distribution, transparent mode

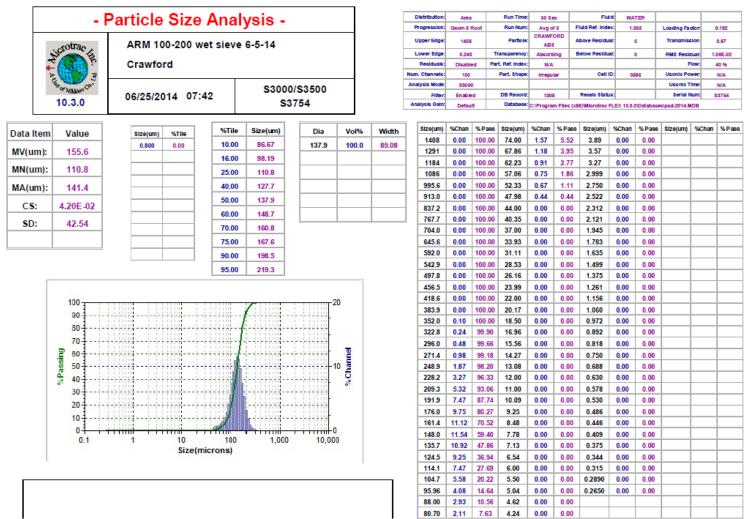


Figure B-11. ARM (-)100/(+)200 mesh, wet sieved, area distribution, absprbamce mode

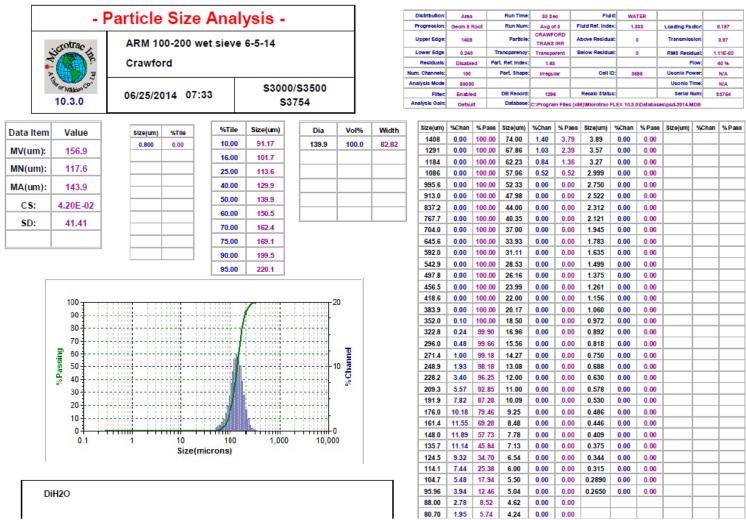


Figure B-12. ARM (-)100/(+)200 mesh, wet sieved, area distribution, transparent mode